Time resolved optical spectroscopy of lead at near critical point states¹.

A. Pyalling^{2,6}, V. Gryaznov², S. Kvitov², D.Nikolaev²,
V. Ternovoi², A.Filimonov², V. Fortov², D.Hoffmann³,
C. Stockl⁴, M. Dornik,⁵

¹Paper presented at the Thirteenth Symposium on Thermophysical Properties, June 22-

^{27, 1997,} Boulder, Colorado, U.S.A.

²Institute of Chemical Physics, Chernogolovka.

³Universitet Erlangen.

⁴Institut fur Angewandte Physik, T H Darmstadt.

⁵Gesellschaft fur Schwerionenforschung, Darmstadt.

⁶To whom correspondence should be wrote.

ABSTRACT

Experiments were performed to investigate the behaviour of lead near its critical point.

Emission spectra of shocked lead samples during unloading into helium at different initial pressures was registered by optical multichannel analyser (OMA), as well as fast optical pyrometer. To describe the obtained experimental data was suggested a model of the thin mixture layer, in which helium emits due to the presence of external electrons from lead.

KEY WORDS: critical state, heat transfer, electron concentration, shock compression, spectroscopy, thermal conductivity.

1.INTRODUCTION

Usage of method of fast multichannel optical pyrometry to study isentropic expansion of the shocked samples allows one to determine pressure of entrance of isentropes into two-phase region [1], to trace the boiling curve and to evaluate parameters of critical point [2]. At the same time, presence of the metal-dielectric transition in this region of phase diagram leads to the increase of emissivity of lead from values 0.3-0.4, usual for liquid metals, up to 1, usual for gases. Heat transfer on the lead-gas boundary also can result in spectral peculiarities, and in different values of temperature in volume and on the boundary. Previous pyrometry experiments gave different brightness temperature for the 585.5 nm channel from the other channels (650, 810, 880, 975 nm). Current spectral measurements were done to investigate registered spectral peculiarities with the aim to improve the pyrometer measurements reliability.

2.MEASUREMENTS

In this work, optical emission from the free surface of shocked and expanded lead samples was registered by means of optical spectroscopy, in spectral range 460-650 nm, with time resolution 20-40 ns, and simultaneously by pyrometer on channels 585.5, 650, 810, 880, 975 nm, with time resolution 3-10 ns. The experimental setup was analogous to [1,3]. Lead samples, shocked up to pressure 265 GPa in the first series of experiments and up to 223 GPa in the second, were unloaded into helium at different initial pressures. Pyrometric measurements with high time resolution were used to determine the temperature of lead in the final state of expansion without heat transfer, by intensity of emission at the moment after sharp rise in the beginning of expansion. Quartz fibbers were used to transfer emission from the experimental setup to the registration apparatus. Velocity of the shock wave in helium was determined by optical base method, permitting

to determine the final pressure of lead expansion by known helium equation of state. A xenon flash lamp was used to determine the spectral sensitivity of spectometer detector assembly. The brightness temperature of the flash lamp was determined by a pyrometer. The pyrometer was calibrated using a tungsten ribbon lamp as a standard source of light.

3.DISCUSSION

The states of lead generated on the expansion isentrope in the first series of experiments corresponds to the non-ideal plasma state, and to the two-phase region state generated after condensation [2]. In the second series of experiments two-phase region states are generating due to an evaporation of the liquid metal on a free surface[3]. The emission spectra, registered after 200 ns from the exit of the shock wave on the free surface of lead are shown in Fig.1. Lead has a lot of emission lines of equal intensity in the investigated spectral range according to spectral data. Line with the wavelength ~590 nm can be clearly seen up to the maximum investigated pressures in the first series of experiments, and at low pressure in the experiments of the second series. This line was not registered in the analogous experiments with lead unloading into hydrogen, but when tin was unloading into helium the same line was observed. So, in the futher analyses it was assumed that this line near 590 nm. corresponds to the helium line 587.6 nm. The time dependence of this line width, its centre position, integral intensity (product of multiplication of width by intensity of line - parameter characterising full probability of line emission) was analysed. For this analysis line shape was approximated by Lorenz form. The misfit of position of the centre of line was about 0.5 nm. The line width was determined with accuracy of ~50%.

Time and spectral dependence of brightness temperature is shown in Fig.2. The integral intensity of this line remains constant during 1 µs, the time needed for a shock wave to

travel through the helium base, with the accuracy of about 10 percent from the initial value.

Time dependence of line position is shown in Fig.3. It can be seen that the position of line and its width remains constant in the initial period of time. After 400-800 ns, the position of line rapidly changes, time of decreasing being about of apparatus resolution. The shift of the helium line from its free atom value was described by the model of electron impact broadening[4]: $\Delta L = \alpha * N_e$, where ΔL - the shift of line position from its value for free atom, N_e - electron concentration, and α - proportionality coefficient. For helium line 587.6 nm theoretical value for α is α =-0.8*10⁻¹⁸ nm *cm³. In the work [5], as well as in our, the opposite sign of this constant was experimentally observed. To evaluate the electron concentration, the theoretical value of α was used, but with the opposite sign. Experimental measured width of line and its position gave N_e -10¹⁸ cm⁻³, changing from 1*10¹⁸ cm⁻³ up to 5*10¹⁸ cm⁻³ with increasing of the final pressure of expansion. Experimental values of N_e from this model are presented in Fig.3 (right axis).

The experimental results were compared with theoretical calculations of the properties of helium and lead performed according to the chemical model of plasma [6]. As initial data for the calculations the velocity of the shock wave in helium with given initial temperature and pressure was taken. Parameters of shocked lead were determined in a separate series of experiments. The shock wave compresses helium up to the state of non-ideal plasma, with mean temperature ~10000 K, and concentration of the electrons $N_e < 10^{16} \text{ cm}^{-3}$. The calculations for lead resulted in a value of $N_e \sim 10^{18}$. cm⁻³. To explain the existence of line on the wavelength 590 nm, it was suggested that helium

emits in a very thin mixture layer, due to the presence of external electrons. This

electrons penetrating into helium from lead. As additional confirmation of this model, the fact may be used that the integral line intensity is constant in time. In the assumption that helium irradiates from the volume, integral intensity must increase in time due to the increase of shocked helium layer width.

The fact that mean position of helium line changes from one value to another, may be explained by arrival of the reflected expansion wave from the inner boundary of lead. Thickness of radiating layer can be estimated from duration of this transition. In present work this time is limited by time resolution of apparatus (20ns). Sound velocity of the shocked helium is about ~5 km/s, so the thickness of the radiating layer is less then 0.1 mm, the result being diminished by order, if lead sound velocity is used for estimation. Time between the exit of the shock wave on the free surface and the moment of the break of line mean position can be used to determine sound velocity in expanded metal. The temperature of radiating layer was evaluated by numerical simulation of heat transfer between two layers (in our case for helium and lead) with a given initial temperatures in volume [7.8]. The volume temperature of lead was obtained from pyrometry measurements and volume temperature of helium was calculated, using chemical plasma model [6]. Heat transfer coefficients were calculated in the first approximation of Chepman-Enskog for the monatomic gas [6]. Electron component of heat transfer was calculated by the equation: $\chi_e = \chi_a \cdot \frac{N_e}{N_a} \cdot \sqrt{\frac{M}{m}}$, where N_e, N_a - concentrations of the electrons and atoms respectively, M, m - masses of atom and electron. Values of Ne, Cv, χ_a , - necessary for the modelling - were obtained from chemical plasma model. It was supposed, that electrons and atoms have equal length of free run [6]. Calculations of temperature on the metal-helium interface were made using standard conductive heat

transfer model [10], taking into account temperature dependence of the temperature conductivity coefficient

Results, obtained from theoretical model and experimental data for four different pressures are presented in Fig.4. Curve 1 is the brightness temperature of helium on the centre of line 587.6 nm, curve 2 - mean spectral temperature of radiating layer (without radiation in lines), curve 3 - temperature of lead obtained by pyrometric measurements, curve 4 - temperature on the helium - lead boundary, calculated from the model of heat transfer, and curve 5 - temperature of shocked helium. One can see, that the calculated temperature on helium-lead boundary obtained from the model of heat transfer does not corresponds to the experimental data.

For the better agreement with the experimental data another model was suggested, in which, it was supposed, that helium and lead are mixed together due to the hydrodynamic instability. As a first approximation it may be assumed, that in this region equal volumes of lead and helium are mixing, and then their temperatures adiabatically equalise. This model results are presented in Fig. 4 (curve 6) and within the experimental error (300 K) agree with experimental points.

Lead electron concentration in mixture layer was calculated according to Griem's dependence of line position as function of electron concentration. Then, lead temperature was calculated for the given pressure and concentration of electrons according to the chemical plasma model [6] (curve 7, Fig.4). Resulting temperatures agree with experimental data for mean spectral temperature at high pressures within experimental error. It is not so at low pressures. It may be supposed that in this case hydrodynamic instabilities at the boundary layer increases. Increase of surface instability can result in entering lead into deeper helium layers, with higher temperatures, so that lead will be ionised at higher initial temperatures.

4.RESULTS

A model of heattransfer was proposed for the boundary between gas and near critical point state metal. This model allows to calculate the volume temperature of expanded metal from the known resultant temperature on the gas - metal boundary. This permits to investigate thermodynamic properties in two-phase region by the pirometres with less time resolution. Also, it was shown that pyrometric measurements have to be performed in spectral interval away from lines of the investigating materials. Spectroscopic measurements also allows to gain more information about the sound velocities in dense nonideal plasma state, and its composition.

ACKNOWLEDGEMENTS

This work was done under financial support by Russian Foundation for Basic Researches (project N 94-02-03706-a) and by INTAS grant No 94-1638.

REFERENCES

- V. G. Ageev, A. V. Bushman, et al., *Pis'ma Zh. Exp. Teor. Fiz.* <u>48</u>:608 (1988) [in Russian].
- V.Ya.Ternovoi, V.E.Fortov, etc.Experimental study of lead critical point parameters.
 Proceedings of 1995 APS Topical Conference "Shock Compression of Condenced Matter".
- Ternovoi V.Ya., Fortov V.E., Filimonov A.S., etc. Experimental study of lead thermodynamics at near critical point parameters, in Proceedings of International Conference on the Physics of Strongly Coupled Plasmas, 1966, World Scientific Publishing Co.Pte.Ltd.
- 4. H.R.Griem, "Spectral line broadening by plasmas.", Academic Press, N. Y. and London 1974.
- 5. H.F.Berg, A.W.Ali, R.Lincke, etc., *Phys.Rev.*, <u>125</u>:199 (1962).
- 6. W.Ebeling, A.Foerster, V.E.Fortov, etc., Thermophysical Properties of Hot Dense Plasmas, Teubner, Stuttgart-Leipzig, 1991.
- 7. W.J.Nellis, H.B.Radousky, etc., *J. Chem. Phys.* 94:2244 (1991).
- 8. J.Ahrens, J.D.Bass, et al., "Shock temperatures in metals", in: SCCM-1989, 1990, pp.851-857.
- 9. A. V. Eleckii, L. A. Palkina, etc., "Yavleniya perenosa v slaboionizirovannoi plasme.", Moscow. Atomizdat. [in Russian]
- 10.R. P. Fedorenko. "<u>Vvedenie v vuichislitelinuyu fiziku</u>. "Moscow, MIPT. 1994. [in Russian].

FIGURE CAPTIONS

- Figure 1. Spectral intensity of emission after 200 ns. from exiting of shock wave into helium.
- Figure 2. Dependece of brightness trmperature versus wavelength and time.
- Figure 3. Position of He (587.6 nm) line center versus time.
- Figure 4. Experimental and theoretical temperatures of helium-lead boundary for different final pressures of expansion.









